

# Orientational order in deposits of magnetic particles

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## Abstract

We present preliminary results for the orientational order in deposits of dipolar particles, on one dimensional substrates. The deposits are generated using a model where the incoming dipolar particle interacts with the other particles in the deposit via a dipole-dipole potential. The inter-dipolar vectors are restricted to lie on a square lattice although the dipole moments are free to rotate in three dimensions. The path of the incoming particle is generated through a Monte Carlo scheme controlled by an effective temperature  $T^*$ , the case of pure diffusion-limited deposition corresponding to  $T^* \rightarrow \infty$ .

We calculate the ferromagnetic and nematic order parameters and the dipolar orientational probability density of the deposits, at various stages of growth and two effective temperatures. The dipolar angular correlations along the rows and columns of the lattice are also investigated. We find that the orientational order of the deposits depends strongly on the lattice structure, the stage of growth and the effective temperature.

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## I. INTRODUCTION

The equilibrium properties of fluids formed by particles carrying strong permanent dipoles have been extensively studied in the last decade [1]. The interest in these systems has grown due to the unusual results obtained in the numerical simulations of dipolar spheres. In fact, almost at the same time, Monte Carlo Canonical (MCC) simulations of dipolar hard spheres in the canonical ensemble [2] and molecular dynamics simulation of dipolar soft spheres [3], have shown that the dipolar fluids exhibit orientational (ferromagnetic) order at densities where the positional order is that typical of a liquid. Moreover, shortly after, MCC simulations revealed that the dipolar fluid at very low densities does not behave as an ideal gas, but instead exhibits a complex structure, with the formation of chains and more complicated polydisperse aggregates [4]. The correlations responsible by these structures have their origin in the anisotropy and long range character of the dipolar potential that become relevant when the temperature is low enough (or the dipole moment is strong enough). These striking results have led to theoretical developments that are capable to describe some of the peculiarities of dipolar fluids (see [1] and references therein).

The dramatic changes obtained for the equilibrium structure of simple fluids when dipolar interactions are taken into account have motivated us to investigate the effects of this interaction in non-equilibrium processes. In this work we present preliminary results obtained when these interactions are included in a model of deposition onto a 1-dimensional substrate.

This paper is organised as follows: in section II we describe our model in detail, and relate it with some of the simplest models of aggregation and deposition as well as with a recent study of aggregation of dipolar particles [5]; in section III we present our preliminary results for the orientational properties of the dipoles in the deposits; finally, in section IV we summarise our findings.

## II. MODEL AND SIMULATION METHOD

One of the simplest models used to study cluster growth is DLA (Diffusion Limited Aggregation) [6]. In this model particles are released, one by one, from a random position at a distance  $R$  from a seed particle, and, after performing a random walk in  $d$ -dimensional

space, attach to the seed or to other particles that already part of the aggregate. DLA was generalized to study the growth of deposits on fibers or surfaces [6, 7]. This generalization is the so called DLD (Diffusion Limited Deposition) model. In DLD particles are deposited onto a  $D$ -dimensional substrate ( $D = 2$  for surfaces and  $D = 1$  for fibers). They diffuse through a  $d = D + 1$  dimensional space, starting at a random position a large distance from the substrate (a plane for  $D = 2$  and a line for  $D = 1$ ), and eventually attach to the substrate or to the deposit formed by previously released particles. These simple models (and modifications thereof) were studied extensively during the last twenty years, with the goal of understanding the formation of complex patterns under conditions far from equilibrium (*e.g.* [6, 8]). A particular extension of DLA was proposed recently in [5] where an off-lattice DLA model is generalized to include dipolar interactions between the particles. The dipolar interactions are taken into account through a Metropolis rule that changes the random diffusive movement of the particles. The interest in deposits of dipolar particles arises from the strongly anisotropic character of the interactions, that may change the fractal dimension of the aggregates, at least under certain conditions [5, 6, 9]. The model used in the present study [9] can be viewed either as a generalization of DLD to include dipolar interactions or as an extension of the dipolar DLA to the growth of deposits on fibers.

The simulations were performed on a (1+1)-dimensional square lattice of width  $L = 800a$  sites and any height to accommodate  $M$  dipoles, where  $a$  is the mesh spacing and the adsorbing substrate coincides with the bottom row (henceforth we take  $a = 1$ ). Periodic boundary conditions are imposed in the direction parallel to the substrate. Each particle carries a 3d dipole moment of strength  $\mu$  and interacts through the pair potential

$$\phi_D(1, 2) = -\frac{\mu^2}{r_{12}^3} [3(\hat{\mu}_1 \cdot \hat{r}_{12})(\hat{\mu}_2 \cdot \hat{r}_{12}) - \hat{\mu}_1 \cdot \hat{\mu}_2], \quad r_{12} \geq a, \quad (1)$$

where  $r_{12}$  is the distance between particles 1 and 2,  $\hat{r}_{12}$  is the two-dimensional (2d) unit vector along the interparticle axis, and  $\hat{\mu}_1$  and  $\hat{\mu}_2$  are the 3d unit vectors in the direction of the dipole moments of particles 1 and 2 respectively. Finally, ‘1’ and ‘2’ denote the full set of positional and orientational coordinates of particles 1 and 2.

A particle is introduced at a lattice site  $(x_{in}, H_{max} + AL)$ , where  $x_{in}$  is a random integer in the interval  $[1, L]$ ,  $H_{max}$  is the maximum distance from the substrate to any particle in the deposit, and  $A$  is a constant. The dipolar moment of the released particle is oriented at random. The particle then undergoes a random walk by a series of jumps to nearest-

neighbour lattice sites, while experiencing the dipole interactions with the particles that are already attached to the deposit. We incorporate the effects of these interactions through the same Metropolis algorithm used in [5]. If the deposit contains  $M$  particles, then the interaction energy of the  $(M+1)$ th incoming particle (the random walker) with the particles in the deposit is given by  $E(\mathbf{r}, \hat{\mu}) = \sum_{i=1}^M \phi_D(i, M+1)$ , where  $\mathbf{r}$  and  $\hat{\mu}$  are the current position and the dipole orientation of the random walker respectively ( $\mathbf{r}$  is a 2d vector). Then we randomly choose a new position  $\mathbf{r}'$  ( $|\mathbf{r} - \mathbf{r}'| = a$ ) and a new dipole orientation  $\hat{\mu}'$  for the random walker [10]; this displacement is accepted with probability

$$p = \min \left\{ 1, \exp \left[ -\frac{E(\mathbf{r}', \hat{\mu}') - E(\mathbf{r}, \hat{\mu})}{T^*} \right] \right\}. \quad (2)$$

$T^* = k_B T a^3 / \mu^2$  is an effective temperature, inversely proportional to the dipolar energy scale. The long range of the dipole-dipole interaction is taken into account in the calculation of the dipolar energy, through the evaluation of the Ewald sums for the particular geometry of this study: 3d dipoles in 2d space with periodic boundary conditions in one direction only [9]. In the limit  $T^* \rightarrow \infty$  all displacements are accepted and our model reduces to DLD. On the other hand, in the limit  $T^* \rightarrow 0$  only displacements that lower the energy  $E(\mathbf{r}, \hat{\mu})$  are accepted. Note that, in this limit, the model still has a certain degree of randomness as the  $(M+1)$ th incoming particle may attach at a site which is not necessarily that corresponding to the absolute minimum of the energy (a lattice row or column).

The diffusion of a particle ends when it contacts the deposit (*i.e.*, becomes a nearest neighbour of another particle that is already part of the deposit) or the substrate (*i.e.*, its position has coordinates  $(x, 1)$ ). After contact, the particle's dipole reorients along the local field due to the other particles in the deposit. When the distance of a particle from the substrate exceeds  $H_{max} + 2AL$ , the particle is removed from the system and another particle is released. In the simulations reported here we took  $A = 1$ ; larger values of  $A$  were tested and found to give the same results, but with drastically increased computation times.

Figure 1 exhibits snapshots of typical deposits obtained at high temperature ( $T^* = 10^{-1}$  in black), and low temperature ( $T^* = 10^{-4}$  in grey). Both deposits have the same general appearance, also observed in DLD: they consist of several trees competing to grow. As the height of the deposit increases, fewer and fewer trees ‘survive’ (*i.e.*, carry on growing), as a consequence of the so-called shielding or screening effect. From figure 1 this seems more pronounced at lower temperatures, since above a height of 1000 (about 1/8 of the maximum

height of this deposit) only one tree survives. The study of the geometrical properties of these deposits was carried out elsewhere [9]. In this paper we will concentrate on the study of the orientational properties of the dipolar deposits.

### III. RESULTS AND DISCUSSION

The orientational properties of the dipoles are studied for deposits at two different effective temperatures,  $T^* = 10^{-1}$  and  $T^* = 10^{-4}$ , for which we simulated 28 and 52 deposits respectively. These values of  $T^*$  were chosen since it is expected that their geometrical properties are close to those of two limiting cases: DLD at the highest temperature,  $T^* = 10^{-1}$ , and  $T^* = 0$  at the lowest,  $T^* = 10^{-4}$  [5, 9].

The study of the global orientational properties of the dipoles as a function of the stage of growth,  $M$ , is carried out through the calculation of the ordering matrix  $\mathbf{Q}$  and the first and second-rank order parameters,  $\langle P_1 \rangle$  and  $\langle P_2 \rangle$ , respectively (see *e.g.* [3]). The mean value of the elements of  $\mathbf{Q}$  when the deposit has  $M$  particles is

$$Q_{\lambda\nu}(M) = \left\langle \frac{1}{2M} \sum_{i=1}^M (3\mu_\lambda(i)\mu_\nu(i) - \delta_{\lambda\nu}) \right\rangle, \quad (3)$$

where  $\lambda$  and  $\nu$  refer to the cartesian coordinates of the unit vector in the direction of the dipole moment and  $\langle \dots \rangle$  stands for an average over all the deposits simulated. The  $x$  and  $y$  coordinates of the dipoles are parallel to the corresponding axes of the underlying square lattice. In figure 2 we plot the values of  $Q_{xx}$ ,  $Q_{yy}$ ,  $Q_{xy}$  and  $Q_{zz}$ .

$Q_{zz}$  approaches  $-\frac{1}{2}$  very rapidly, corresponding to the vanishing of the out of plane component of the dipole moment,  $\mu_z$ : except for the first hundreds of particles, the dipoles lie on the plane of the deposit. This property resembles that of 2d dipolar fluids [11] and is due to the anisotropy of the dipolar potential, that enforces the dipoles to lie on the plane of the interdipolar vectors.

There is a very fast transient ( $M < 30 - 100$  depending on  $T^*$ ) corresponding to deposits with a finite out of plane dipole moment. In this initial regime  $Q_{zz}$  decreases rapidly while  $Q_{xx}$  increases and  $Q_{yy}$  decreases. Although the number of deposited particles is rather small the absolute value of the  $xx$  component of  $\mathbf{Q}$  is systematically larger than that of the  $yy$  component suggesting an anisotropic initial growth.

We then distinguish an early stage ( $30 < M < 100$  at  $T^* = 10^{-4}$  and  $100 < M < 3000$

at  $T^* = 10^{-1}$ ) where  $Q_{xx}$  decreases while  $Q_{yy}$  increases steeply. This is followed by a late stage characterized by slow increase of  $Q_{xx}$  and a decrease of  $Q_{yy}$  to their asymptotic values of 0.25 (note that the horizontal scale of figure 2 is logarithmic).  $Q_{xx}$  exhibits a minimum at a value of  $M$  that depends on temperature and  $Q_{yy}$  has the opposite behavior with a maximum at (roughly) the same value of  $M$ . The difference between these extrema decreases as  $T$  increases.

The value of  $Q_{xy}$  is  $\approx 0$  for the entire deposit (even in the initial stage) at both effective temperatures. In fact, all the other non-diagonal elements,  $Q_{\lambda\nu}$  with  $\lambda \neq \nu$ , also vanish and the ordering matrix is diagonal (coupling of the lattice and dipolar potential anisotropies).

These results for  $\mathbf{Q}$  imply that the orientation of the dipoles, after the first hundreds of particles are released, is restricted to the plane of the deposit and is preferentially along one of the two lattice directions: parallel to the substrate, at the initial stage of growth, and perpendicular to it at the early stage. These preferred directions result from the coupling between the lattice and potential anisotropies: nearest neighbours on the lattice have horizontal or vertical interdipolar vectors and the lowest energy of a dipole pair corresponds to dipoles aligned along the interdipolar vector. Finally at the late stage of growth, the two lattice directions are (almost) equivalent.

The order parameters  $\langle P_1 \rangle$  and  $\langle P_2 \rangle$  provide information about the ferromagnetic and nematic order of the system. In the simulations of liquid crystalline systems [12] and dipolar fluids [3],  $\langle P_2 \rangle$  is identified with the the largest eigenvalue of the ordering matrix,  $\Lambda_{max}$ . This definition is appropriate when the other eigenvalues are  $\approx -\frac{1}{2}\Lambda_{max}$  [12]. As is clear from the results for  $Q_{\lambda\nu}$ , this condition is not fulfilled in the present case. For instance, in the late stage of growth,  $Q_{xx} \approx Q_{yy} \approx \frac{1}{4}$ ,  $Q_{zz} \approx -\frac{1}{2}$  and  $Q_{\lambda\nu} \approx 0$  if  $\lambda \neq \nu$ . The eigenvalues ( $\Lambda_{max}, \Lambda_0, \Lambda_{min}$ ) are then  $(\frac{1}{4}, \frac{1}{4}, -\frac{1}{2})$ , meaning that the dipoles have a vanishing component along one direction (perpendicular to the plane of the deposit) and that their orientations are distributed symmetrically along the other two perpendicular directions (within the plane of the deposit). Thus, the dipoles are “randomly” oriented in the plane of the deposit and the system has no orientational order, apart from that imposed by the underlying lattice. This suggests an alternative definition of  $\langle P_2 \rangle$  as the difference between  $\Lambda_{max}(M)$  and the maximum eigenvalue of  $\mathbf{Q}$  obtained for a deposit of  $M$  dipoles, randomly oriented on a plane,  $\Lambda_{max}^{ran}(M)$ . The mean value of  $\langle P_2 \rangle$  at each stage of the growth  $M$  is then calculated

from,

$$\langle P_2(M) \rangle = \langle \Lambda_{max}(M) \rangle - \langle \Lambda_{max}^{ran}(M) \rangle, \quad (4)$$

where  $\langle \Lambda_{max}^{ran}(M) \rangle$  was calculated using 100 configurations of 50000 random dipoles.

Figure 3a shows the results for  $\langle P_2 \rangle$  at two temperatures. The nematic order parameter has the same qualitative behavior at the two temperatures. At  $T^* = 10^{-1}$  during the initial stage of growth  $\langle P_2 \rangle$  increases abruptly from zero to about 0.5 ( $M < 100$ ). In the late stage ( $M > 3000$ )  $\langle P_2 \rangle$  oscillates around a small value ( $< 0.1$ ) and then decreases slowly to zero as  $M \rightarrow \infty$ . The early stage ( $100 < M < 3000$ ) is dominated by a marked decrease of  $\langle P_2 \rangle$  that goes almost to zero and then increases to a value below 0.1. The behaviour of  $\langle P_2 \rangle$  at the lower temperature is very similar.

Taking into account the results obtained for the elements of the ordering matrix  $\mathbf{Q}$  this means that at the initial stage the dipoles have a tendency to point along the lattice direction parallel to the substrate; at the early stage there is a tendency to align vertically; and finally, at the late stage of growth, there is a tendency to align both vertically and horizontally with (almost) equal probability. There are, however, quantitative differences between the behavior at the two temperatures. The tendency to align horizontally at the initial stage decreases with decreasing temperature, since as  $T$  decreases it occurs for smaller deposits and is characterized by a lower value of  $\langle P_2 \rangle$ . On the other hand, the tendency to align vertically at the early stage is enhanced at low temperatures since as  $T$  decreases the value of the second maximum of  $\langle P_2 \rangle$  increases and the decay towards the late stage behavior is much slower.

The ferromagnetic order parameter  $\langle P_1 \rangle$  is defined, for equilibrium systems of dipolar particles, as the projection of the polarization onto the director of the system [3]. The director,  $\hat{d}$ , is the eigenvector of  $\mathbf{Q}$  corresponding to  $\Lambda_{max}$ . Therefore, we calculate the ferromagnetic order parameter using,

$$\langle P_1(M) \rangle = \frac{1}{M} \left\langle \left| \sum_{i=1}^M \hat{\mu}_i \cdot \hat{d}(M) \right| \right\rangle. \quad (5)$$

Figure 3b shows the results for  $\langle P_1(M) \rangle$  at two temperatures. In both cases  $\langle P_1(M) \rangle$  reaches a maximum at the end of the initial stage of growth and then falls rapidly in the early stage. At the higher temperature  $\langle P_1 \rangle$  is almost zero during the late stage of growth. At the lower temperature, however,  $\langle P_1 \rangle$  is non-zero in the late stage of growth with a broad maximum ( $\sim 0.15$ ) at  $M \sim 10000$ . For larger  $M$   $\langle P_1 \rangle$  exhibits a slight tendency to

decrease but from these results it is not clear that the asymptotic value of the ferromagnetic order parameter vanishes as  $M \rightarrow \infty$ . If we consider the results obtained previously for the elements of the ordering matrix, we conclude that the increase in ferromagnetic order for  $M > 300$  (extending from the initial to the late stage regimes) corresponds to dipoles aligned along the vertical direction.

We proceed this preliminary analysis by calculating the orientational probability density (OPD) as a function of the stage of growth,  $P(\theta, M)$ .  $P(\theta, M)d\theta$  is the probability of finding two dipoles with a relative orientation between  $\theta$  and  $\theta + d\theta$ , in a deposit with  $M$  dipoles. This is calculated by dividing  $[0; \pi]$  into intervals  $\Delta\theta$ , and counting the number of pairs of dipoles  $n(\theta_k, M)$ , with relative orientation in the range  $]\theta_k; \theta_k + \Delta\theta[$ , for a deposit with  $M$  dipoles. The OPD is given by

$$P(\theta_k, M) = \frac{2}{M(M-1)} \frac{\langle n(\theta_k, M) \rangle}{\Delta\theta} \quad (6)$$

In figure 4 we plot the OPD at three stages of growth,  $M = 2000, 10000$  and  $50000$ , for  $\Delta\theta = \pi/100$ . If the relative orientation of the dipoles were random the OPD would be uniform ( $1/\pi$ ). Our results indicate a highly non-uniform behaviour. The peaks, at  $0, \pi, \pi/2, \pi/4$  and  $3\pi/4$ , occur at both temperatures, at every stage of growth. At the lower temperature, the structure is enhanced and other peaks appear. The two large peaks at  $0$  and  $\pi$  indicate that most dipoles are aligned while the peak at  $\pi/2$  shows that the alignment occurs along two perpendicular directions. At first sight, the OPD appears to be symmetric about  $\pi/2$ . A more careful analysis, however, indicates a difference in the heights of the peaks at  $0$  and  $\pi$ , that is largest in the early stage of growth and at the lowest temperature: the peak at  $\pi$  is lower than that at  $0$ . This indicates that the system exhibits ferromagnetic order. Consider an off-lattice system where the OPD may be approximated as  $\langle P(\theta, M) \rangle \approx \frac{1}{\pi} (1 + A_1 P_1(\cos \theta) + A_2 P_2(\cos \theta))$ , with the coefficients  $A_i$  proportional to the square of the order parameters  $\langle P_i(M) \rangle$ . This function has a maximum at  $0$ . If  $A_1 = 0$  and  $A_2 \neq 0$  then it is symmetric about  $\pi/2$ ; if,  $A_1 \neq 0$  the function has either a minimum or a (local) maximum at  $\pi$ , depending on the values of  $A_i$ ; but, in any case, it is not symmetric about  $\pi/2$ . The difference between the values of the OPD at  $0$  and  $\pi$  is proportional to  $\langle P_1 \rangle^2$ . We see from figure 4 that, although in the present case the OPD exhibits a lot more structure due to the underlying lattice, the observed asymmetry of the peaks at  $0$  and  $\pi$  is consistent with the results for  $\langle P_1 \rangle$ : at  $T^* = 10^{-1}$  the asymmetry is apparent at the early



stage of growth, *i.e.*, for  $M = 2000$  only, while at  $T^* = 10^{-4}$  the asymmetry is evident for all stages of growth.

The results for the OPD and the order parameter  $\langle P_1 \rangle$  may be compared with the results of [5]. In this off-lattice study of DLA-dipolar clusters, the OPD was calculated and the function  $\frac{1}{\pi}(1 + A_1(T^*) \cos \theta)$  was shown to be a good approximation to it. At the temperatures considered in this work the values  $A_1(10^{-1}) \approx 0$  and  $A_1(10^{-4}) \approx 0.05$  were obtained in [5]. Thus, our results are consistent with those of [5]: both dipolar DLA clusters and dipolar DLD deposits become polarized below a certain temperature, at least for values of  $M \sim 50000$ .

To illustrate the relation between the results obtained so far and the spatial properties of the deposits and to motivate our last calculation, we plot in figure 5 two snapshots at  $T^* = 10^{-4}$  and  $T^* = 10^{-1}$  (dipoles with  $|\hat{\mu}_x|/|\hat{\mu}_y| < 1$  ( $> 1$ ) are plotted in black (grey)). These structures are formed mostly by horizontal grey rows and vertical black columns, suggesting that the particles have a tendency to form chains, along the lattice directions. Elsewhere, the connection between the spatial and the orientational order was verified quantitatively [9]. Briefly, it was shown that the orientation of a dipole on the substrate has a tendency (that increases with decreasing temperature) to be in the direction of growth of the deposit at the site where the dipole was attached. Here, we want to quantify the relation between the orientation of a pair of dipoles and their distance along a chain, at different temperatures  $T^*$ . Since the dipoles are oriented along the rows and columns of the underlying lattice, in a head to tail alignment, an estimate of the average size of the dipolar chains is given by the distance beyond which the orientational correlations vanish. For a given deposit we calculate the average value of the cosine of the angle between all pairs of dipoles along rows (or columns) that are a given distance apart. For columns this is

$$\langle \cos \alpha(r) \rangle_v = \left\langle \sum_{i=1}^L \sum_{j=1}^{h^*(i)} \frac{\hat{\mu}(i, j) \cdot \hat{\mu}(i, j+r)}{n_d(i, r)} \right\rangle, \quad (7)$$

where  $\hat{\mu}(i, j)$  is the dipolar unit vector at site  $(i, j)$ ,  $h^*(i)$  is the maximum height of column  $i$  and  $n_d(i, r)$  is the number of pairs of dipoles in column  $i$  a distance  $r$  apart. An analogous calculation for rows is,

$$\langle \cos \alpha(r) \rangle_h = \left\langle \sum_{j=1}^{h^*} \sum_{i=1}^L \frac{\hat{\mu}(i, j) \cdot \hat{\mu}(i+r, j)}{n_d(j, r)} \right\rangle. \quad (8)$$

where  $h^*$  is the maximum height of the deposit,  $n_d(j, r)$  is the number of pairs of dipoles

in row  $j$  a distance  $r$  apart. Periodic boundary conditions in the lateral direction are taken into account.

Figure 6 shows  $\langle \cos \alpha(r) \rangle_v$  and  $\langle \cos \alpha(r) \rangle_h$  at the late stage of growth ( $M = 50000$ ). At short distances, the curves exhibit an oscillatory behaviour, along rows and columns, at both temperatures. At intermediate distances the curves decrease exponentially and at large distances they oscillate around zero (for the distances depicted in figure 6, this is seen only for the horizontal case at  $T^* = 10^{-1}$ ), corresponding to the loss of correlation between the dipolar orientations.

The oscillatory behavior at short distances is due to the interplay between the anisotropy of the dipolar interaction and the lattice. Suppose that a dipole makes an angle  $\alpha$  with *e.g.* the vertical direction. If another dipole attaches to the deposit as a top neighbour, the angle with the vertical that minimizes the interaction energy between the two dipoles is  $-\alpha$  (neglecting the interactions with the other dipoles in the deposit). It then follows that the third, fifth, *etc.*, dipoles align at an angle  $\alpha = 0$  with the first giving rise to the (damped) oscillations observed at short distances ( $r < 5 - 20$  lattice spacings).

The decay of angular correlations, for rows and columns, is temperature dependent. At fixed  $r$ ,  $\langle \cos \alpha(r) \rangle_v$  and  $\langle \cos \alpha(r) \rangle_h$ , are larger at the lower temperature, since the tendency for dipolar alignment increases as the temperature is lowered. In the inset of figure 6 we have plotted  $\langle \cos \alpha(r) \rangle_v$  and  $\langle \cos \alpha(r) \rangle_h$  in a logarithmic scale. The plots are linear indicating that the correlations decay exponentially. The differences are not large, but the decay of correlations is faster at higher temperatures (for rows and columns) and slower for columns (at any fixed temperature). We conclude that the dipoles are aligned in columns or rows forming chains with a typical size, that depends on temperature. A similar analysis at the early stage of growth will probably reveal larger differences.

#### IV. CONCLUDING REMARKS

From the results for the order parameters and the ordering matrix  $\mathbf{Q}$  we conclude that the dipoles of the deposits have a slight tendency to align parallel to the substrate at the initial stage and perpendicular to it during the early stage of growth. The vertical (horizontal) alignment is enhanced by decreasing (increasing) temperatures. We have found evidence that the dipoles exhibit ferromagnetic order, at low temperatures, in agreement with the

results of the off-lattice dipolar DLA model [5]. However, we cannot discard the possibility that this ferromagnetic order is a finite size effect: in fact, at  $T^* = 10^{-4}$  the mean height of the deposit for  $M = 50000$  is almost one order of magnitude larger than that at  $T^* = 10^{-1}$  [9].

The fact that the nematic director is always either horizontal or vertical is of course a lattice effect; but the existence of a temperature dependent nematic order that changes from parallel to the substrate (in the initial) to perpendicular (in the early stage of growth), seems more likely to occur in off-lattice versions of this model.

Finally, we have shown that the chained structure of the deposits has a characteristic length that depends on the direction of the chains and on the temperature. The relation between the orientational order studied in this work and the geometrical properties (fractal dimension) of the deposits [9] is left for future work.

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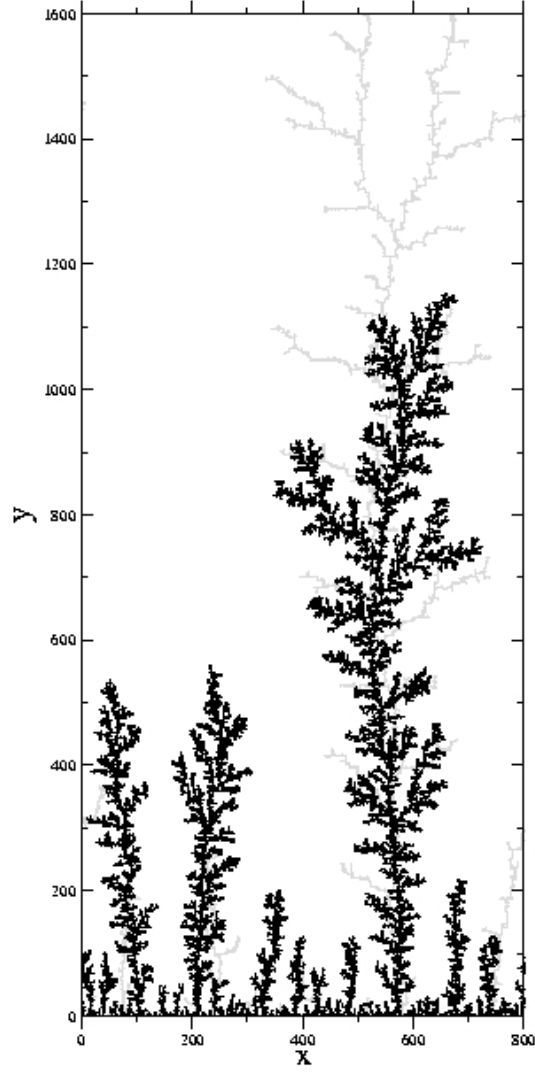


FIG. 1: Snapshots of two deposits at  $T^* = 10^{-1}$  (black) and  $T^* = 10^{-4}$  (grey).

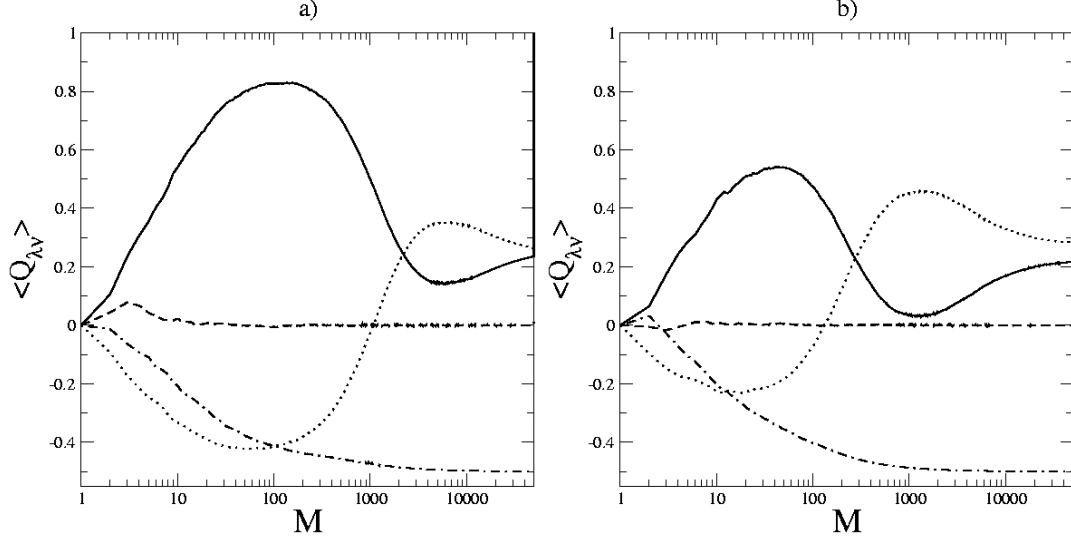


FIG. 2: Mean values of the components of the ordering matrix  $\mathbf{Q}$  as a function of the stage of growth. We identify three regimes: (i) initial growth with decreasing  $Q_{zz} \neq -0.5$ , (ii) early growth with decreasing  $Q_{zz} \sim -0.5$  and increasing  $Q_{xx}$  and decreasing  $Q_{yy}$  and (iii) late growth with  $Q_{zz} = -0.5$  and increasing  $Q_{xx} \sim$  decreasing  $Q_{yy}$  (see text for details). (a),  $T^* = 10^{-1}$  and (b),  $T^* = 10^{-4}$ .  $Q_{xx}$  - full line;  $Q_{yy}$  - dotted line;  $Q_{zz}$  - dot-dashed line;  $Q_{xy}$  - dashed line.

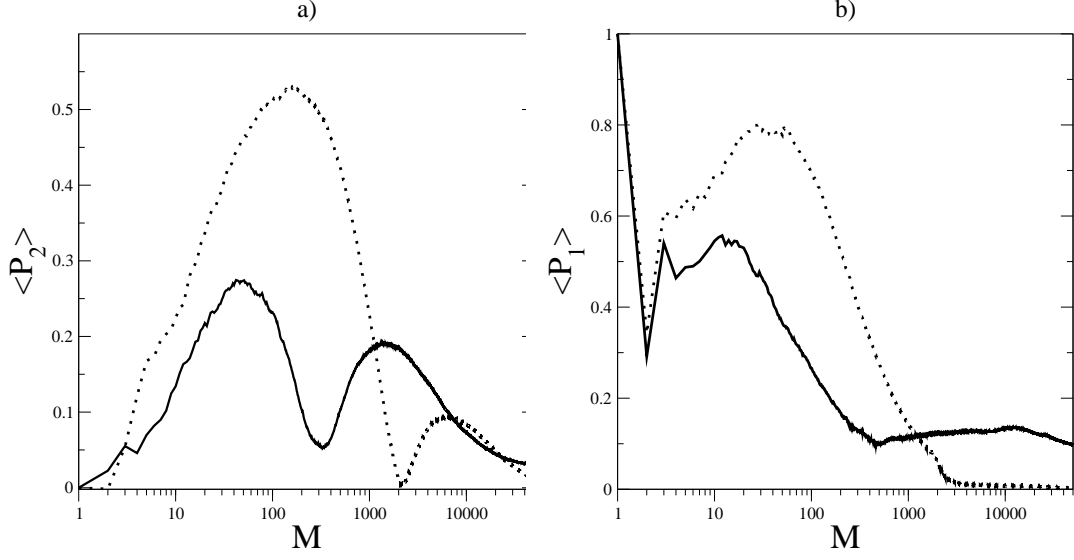


FIG. 3: Mean values of (a) the nematic ( $\langle P_2 \rangle$ ) and (b) the ferromagnetic ( $\langle P_1 \rangle$ ) order parameters as a function of the stage of growth. Dotted line:  $T^* = 10^{-4}$ . Full line:  $T^* = 10^{-4}$ .

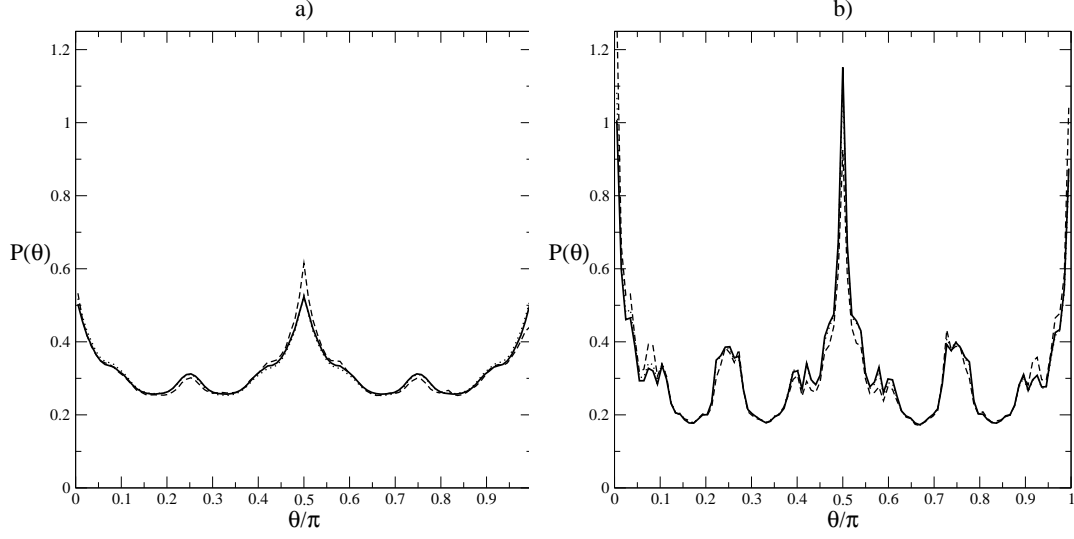


FIG. 4: Mean value of the orientational probability density at  $T^* = 10^{-1}$  (a) and at  $T^* = 10^{-4}$  (b). The different lines correspond to different sizes of the deposit: full line,  $M = 50000$ ; dotted line  $M = 10000$ ; dashed line  $M = 2000$ .



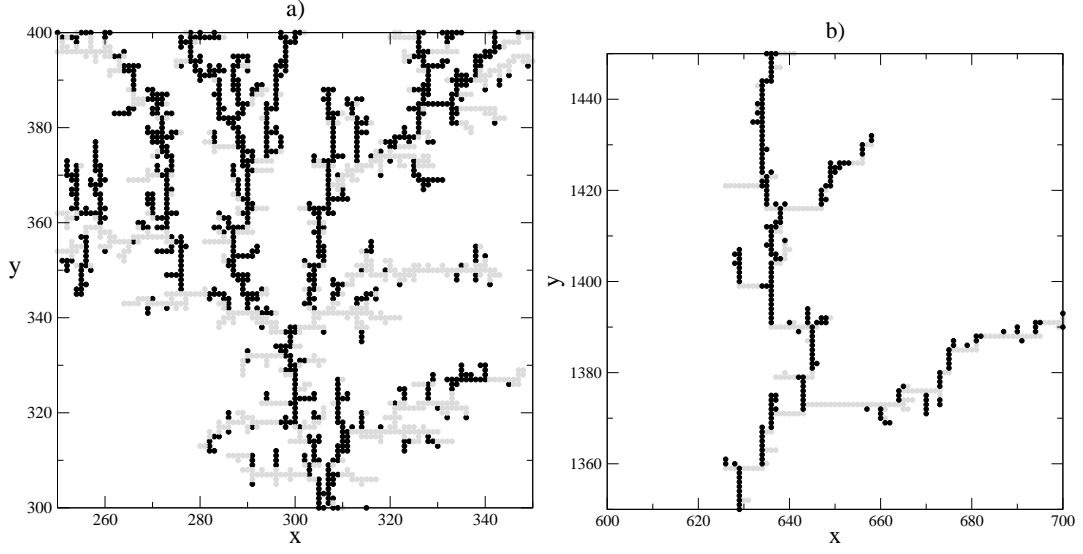


FIG. 5: Details of snapshots for two deposits at  $T^* = 10^{-1}$  (a) and at  $T^* = 10^{-4}$  (b). The dipoles whose absolute value of the vertical component is larger (smaller) than the absolute value of the horizontal component are plotted in black (grey).

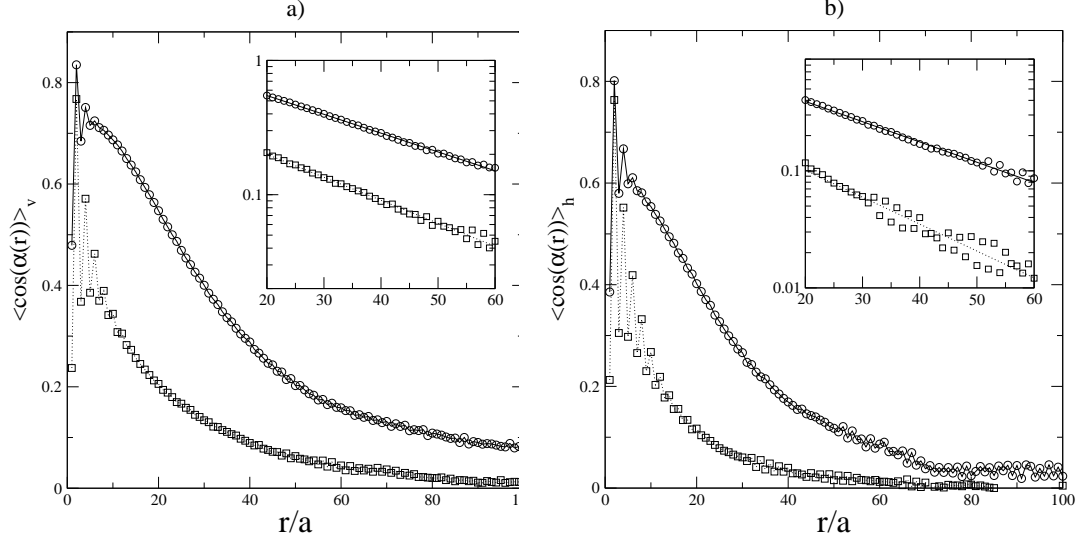


FIG. 6: Mean value of the co-sine of the angle between dipoles on the same column (a) or row (b) a distance  $r$  apart for late stage deposits, with  $M = 50000$ . The circles correspond to  $T^* = 10^{-4}$  and the squares to  $T^* = 10^{-1}$ . The lines in the main figures are a guide to the eye. The lines in the insets are linear regressions illustrating the exponential decay of correlations in that range of distances.